Quasi-Stationary Measurements of Lignamon Thermal Properties

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Thermal properties of wood and modified wood-based materials are important parameters that influence the manufacturing process and final industrial utilization. The aim of this work was to investigate three main thermal properties (thermal conductivity, thermal diffusivity, and specific heat capacity) of ammonia-treated compressed beech wood (Lignamon material) and natural beech wood (*Fagus sylvatica*).These properties were measured based on the quasi-stationary method developed at the Department of Wood Science at the Technical University in Zvolen. The influence of increased density (caused by ammonium treatment and compression) of four different types of Lignamon material on the thermal properties was discovered, and the results were compared with those from untreated beech wood. The results confirmed a dependency on the density of the material. With increasing Lignamon compression extent (increasing density value), the thermal conductivity increased and the thermal diffusivity decreased. The maximum value of thermal conductivity reached (0.26 $W.m^{-1}.K^{-1}$ at 1070 $kg.m^{-3}$) in the case of Lignamon 6k and (0.26 $W.m^{-1}.K^{-1}$ at 950 $kg.m^{-3}$) in the case of Lignamon 7n.

Keywords: Lignamon; Thermal conductivity; Specific heat capacity; Thermal diffusivity; Quasi-stationary method

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INTRODUCTION

The substitution of improved domestic species for unavailable high-grade tropical wooden species has led to progress in modification processes. The current state of applied wood modifications is described in the work of Hill (2006) and Rowell (2006). Modifications of wood from local sources are still very topical (Hill 2011; Esteves and Pereira 2009), and the use of modern technologies enables outdated ideas to become more widely used, efficient, and environment-friendly. Wood modification has been viewed as the right approach to achieve the desired properties of wood with a minimum of expended energy and materials.

Interestingly, ammonia-treated wood was developed in Czech lands in the early 1960s (Czerny and Valasek 1974; Berzins 1972; Davidson and Baumgardt 1970; Kalnins *et al.* 1967; Pandey *et al.* 1991). This material is called Lignamon, a compressed beech wood modified by ammonium. The technological process is composed of four basic steps. First, untreated beech wood with a high moisture content (18 ± 3%) is heated to a maximum of 103 °C. Second, the sample is plasticized using ammonia steam. The process of modification is discontinuous. Then, the sample is compressed within the range of 0.8 to 1.3 MPa. The higher the pressure is, the denser the final Lignamon sample becomes. After disposing of the spare nitrogen from the modified sample, the drying and stabilization procedures are performed. Lignamon exhibits higher durability and density and darker colour, as recently reported by Weigl *et al.* (2009, 2012). Lignamon has been used in mechanical engineering, furniture, and musical instrument production. Lignamon can also be used for many other purposes: it can be used as a substitute for imported tropical wood, alloys, and plastics, and as material for handgrips, shafts, flooring, and bearing surfaces (Sedliačik 1998).

The properties of Lignamon are primarily based on the properties of raw wood material (*Fagus sylvatica* L.) and its technological processing. The process of transforming the native material into Lignamon influences the density, mechanical properties, and appearance (Stojčev 1979). The least mentioned and examined properties of Lignamon are thermal conductivity, thermal diffusivity, and specific heat capacity.

There are two different methods (stationary and non-stationary) for measuring the thermal properties of materials; these methods have been described by many authors. The thermal conductivity measurements of beech wood with the stationary method (guarded hot plate) have been carried out by Niemz *et al.* (2010), MacLean (1941), Skaar (1988), and others. The wood species itself has no important effect on thermal conductivity, which means that data from other sources for almost any species with a known specific gravity can be compared (Steinhagen 1977). The unsteady state method has been used to obtain all three thermal properties of different wooden species. Hrčka (2010) employed a method that contacted air at the beech wood surface to establish the thermal properties. Transient measurements of thermal conductivity and thermal diffusivity using the plane source technique on hardwood samples were introduced by Suleiman *et al.* (1999). Kühlmann (1962) investigated the thermal properties of spruce, oak, and beech with the Krischer and Esdorn apparatus. A nonsteady-state heating device was used to determine the thermal properties of larch (Chudinov and Stepanov 1971).

The quasi-stationary method employed in this work was developed at the Department of Wood Science at the Technical University in Zvolen (Požgaj *et al.* 1997). The method is based on two principles described by Clarke and Kingston (1950) and Krisher and Esdorn (1954). Quasi-stationary is defined as resembling stationary conditions resulting from the linear part of the temperature measurement needed for the evaluation of thermal properties.

The main goal of this article was to establish the thermal properties (thermal conductivity, thermal diffusivity, and specific heat capacity) of Lignamon material and untreated beech wood (UB) using the quasi-stationary method. The specific objectives were as follows: a) to investigate the influence of increased density (measured by X-ray densitometer) of Lignamon on the thermal properties, and b) to establish the accuracy of the measuring method and compare the results for native beech wood with the available literature sources.

EXPERIMENTAL

There are two main parts to the present research: a) material preparation, evaluation of density profiles, air conditioning of samples, and experimental measurements of thermal properties; and b) the processing of data according to the least squares method using a macro written in Excel (Hrčka and Babiak 2012). Four different types of Lignamon samples were cut from massive blocks (Lignamon 6k and 7n) or thin planks

(Lignamon 24 and 25) with approximate dimensions of 50 x 50 x 8 mm (width x length x thickness). The wood fiber directions are longitudinal in width, radial in length, and radial-tangential in thickness.

Thermal properties were measured in the direction of heat flow, which was the radial-tangential direction for all samples. Samples of Lignamon 6k and 7n were highly pressed (compression extent of 38%) during the modification process and therefore reached higher densities and darker colors. Lignamon 24 and 25 were made of varnished floor parquets and had brighter colors. Eight samples were made from each type of Lignamon. Next, eight samples of untreated beech (UB) wood were prepared to compare the thermal properties of Lignamon with unmodified wood samples. An X-ray densitometer (X-RAY Dense-Lab, Germany) was used to scan all samples with a scanning step length of 0.01 mm. An average density profile from each type of sample was established. All 40 samples were conditioned at 20 °C and a relative humidity of 65% in a Sanyo MTH 2400 air chamber to reach the equilibrium moisture content.

The non-stationary method (quasi-stationary) was based on the evaluation of temperature changes in measured samples. The theory's background lies in solving the heat transfer equation with specific boundary conditions (constant heat flux on the upper surface of the measured sample, zero heat flow in the middle of the apparatus reached due to the sample's symmetry, and a constant temperature in the whole sample) (Fig. 1). The apparatus is composed of eight samples that are alternately interspersed with NiCr thin foil (0.01 mm) to produce heat. The block of samples is insulated by polystyrene and pertinax on both sides of the apparatus. A rise in temperature was measured in the middle of the samples and was recorded with a thermocouple and a computer (Požgaj *et al.* 1997; Hrčka and Babiak 2012). The average heat flux generated by the heat source and entering the samples through the thin foil was in the range of 957 to 1052 W/m^2 for different measurements. Together, 13 different sample arrangements of each material type were measured. Thermal properties were established as approximate values of two samples placed in the center of the apparatus, between which the temperature sensor was placed. After each measurement, the two middle samples were changed and their average densities and thermal properties evaluated.

The data from the measurements are illustrated in Fig. 2 as a curve and describe the temperature increase in time in the center of the middle sample.

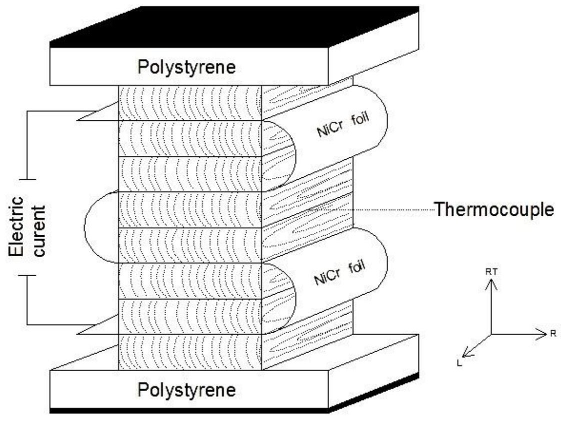

Fig. 1. Scheme of the apparatus

There were two nonlinear parts of the curve. The beginning part could be expressed as the sum of an infinite series, and the other part represents the end of the measurement (caused by heat loss through lateral surfaces). The middle part was linear, as is the temperature increased with time. The thermal properties were calculated by interpolation of the curve using the theoretical line (programmed in Visual Basic for Applications).

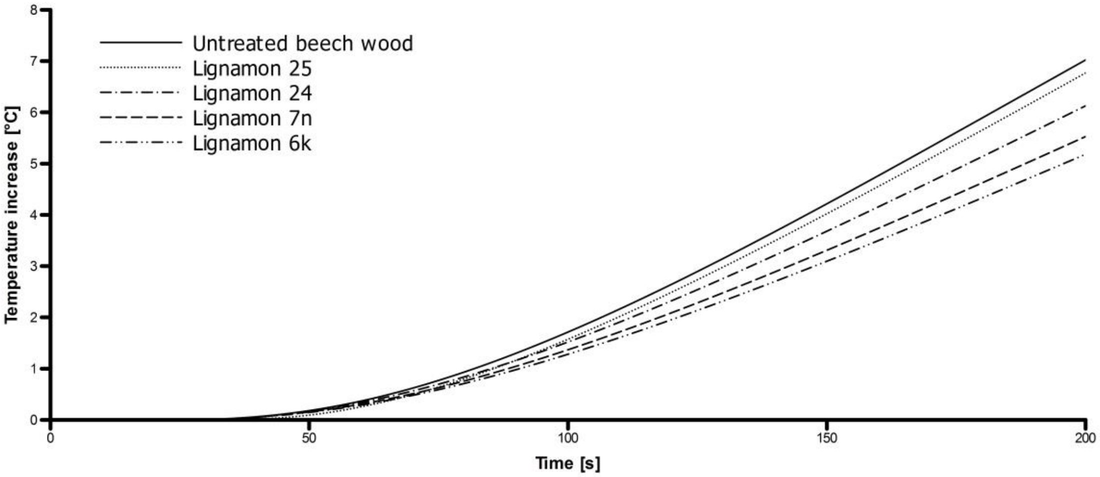

Fig. 2. The temperature increase between central samples recorded by a thermocouple

RESULTS AND DISCUSSION

Density profiles of all samples reached almost uniform distribution, with no large fluctuations. Only Lignamon 25 exhibited a slight decrease in density in the middle of the samples. X-ray densitometry results are shown in Fig. 3. The lower density values from surface layers with depth of 0.1 mm were caused by the roughness and non-flatness of surfaces. This so-called edge effect originated during preparation of samples and influenced the scanning process. The differences between microstructure of untreated beech wood and compressed Lignamon are shown in Fig. 4. The densification process caused deformation of the wood structure, especially buckling from fibers and change in growth rings orientation.

The thermal conductivity, thermal diffusivity, and specific heat capacity were derived from the measurements based on the quasi-stationary method. The results are presented in Table 1. The measured data confirm the dependence on density (compression rate of ammonium-treated samples), as shown in Fig. 5. In general, thermal conductivity increases as the density, moisture content, or temperature of the wood increases (see Kühlmann 1962). The more the Lignamon samples were pressed (Lignamon 6k and 7n), the higher the density and thermal conductivity values became. However, the dependence of thermal diffusivity on the density value followed an inverted trend.

The specific heat capacity of wood is practically independent of density, which was confirmed by our measurements. The average equilibrium moisture content (7.8 % with standard deviation 0.5) of all measured samples was established according to the gravimetric method.

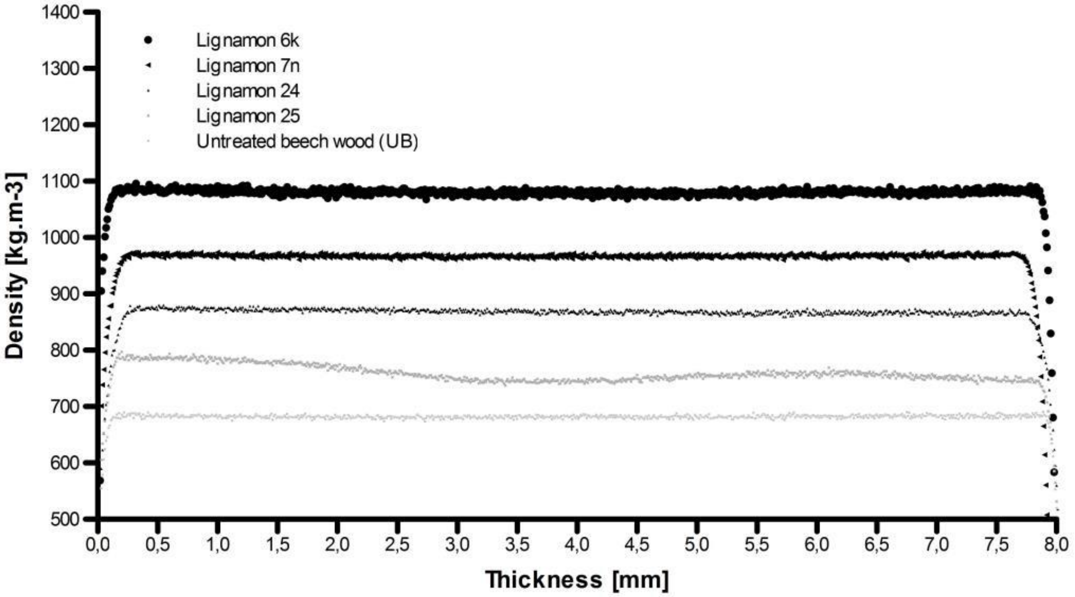

Fig. 3. Average density profiles of all samples (mean coefficients of variation in %: Lignamon 6k- 1.8; Lignamon 7n- 3.4; Lignamon 24- 2.0; Lignamon 25- 2.8; UB- 1.6)

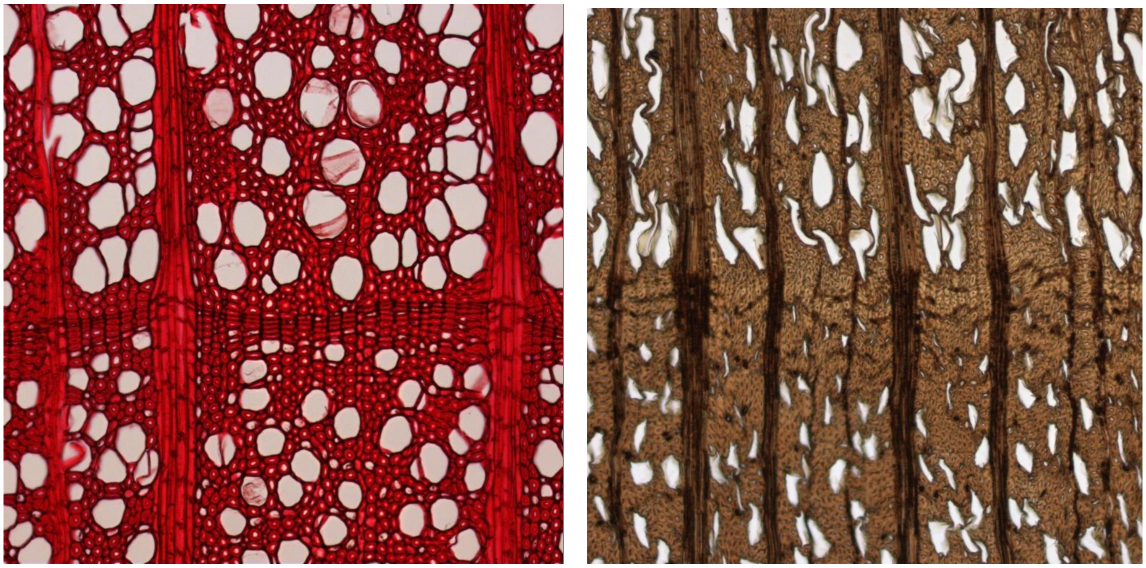

Fig. 4. Microstructure pictures of untreated beech wood (on the left side) and Lignamon (on the right side); magnification of approximately 100x

Table 1. Mean Properties of Samples Measured in Radial-Tangential Direction

Property	UB (untreated beech wood)	Lignamon 25	Lignamon 24	Lignamon 7n	Lignamon 6k
ρ [kg.m^{-3}]	675 (4.5)	753 (11.7)	857 (8.0)	950 (18.9)	1070 (9.4)
λ [W.m^{-1}.K^{-1}]	0.21 (0.05)	0.21 (0.07)	0.25 (0.04)	0.26 (0.06)	0.26 (0.05)
a [10^{7} m^{2}.s^{-1}]	1.2 (0.2)	1.2 (0.2)	1.2 (0.2)	1.1 (0.2)	1.1 (0.1)
c [J.kg^{-1}.K^{-1}]	2632 (272)	2350 (395)	2397 (212)	2519 (314)	2244 (234)
Mean: density (ρ), thermal conductivity (λ), thermal diffusivity (a), and specific heat capacity (c) for 7.8% of EMC; the values in parentheses describe the standard deviation of measurement					

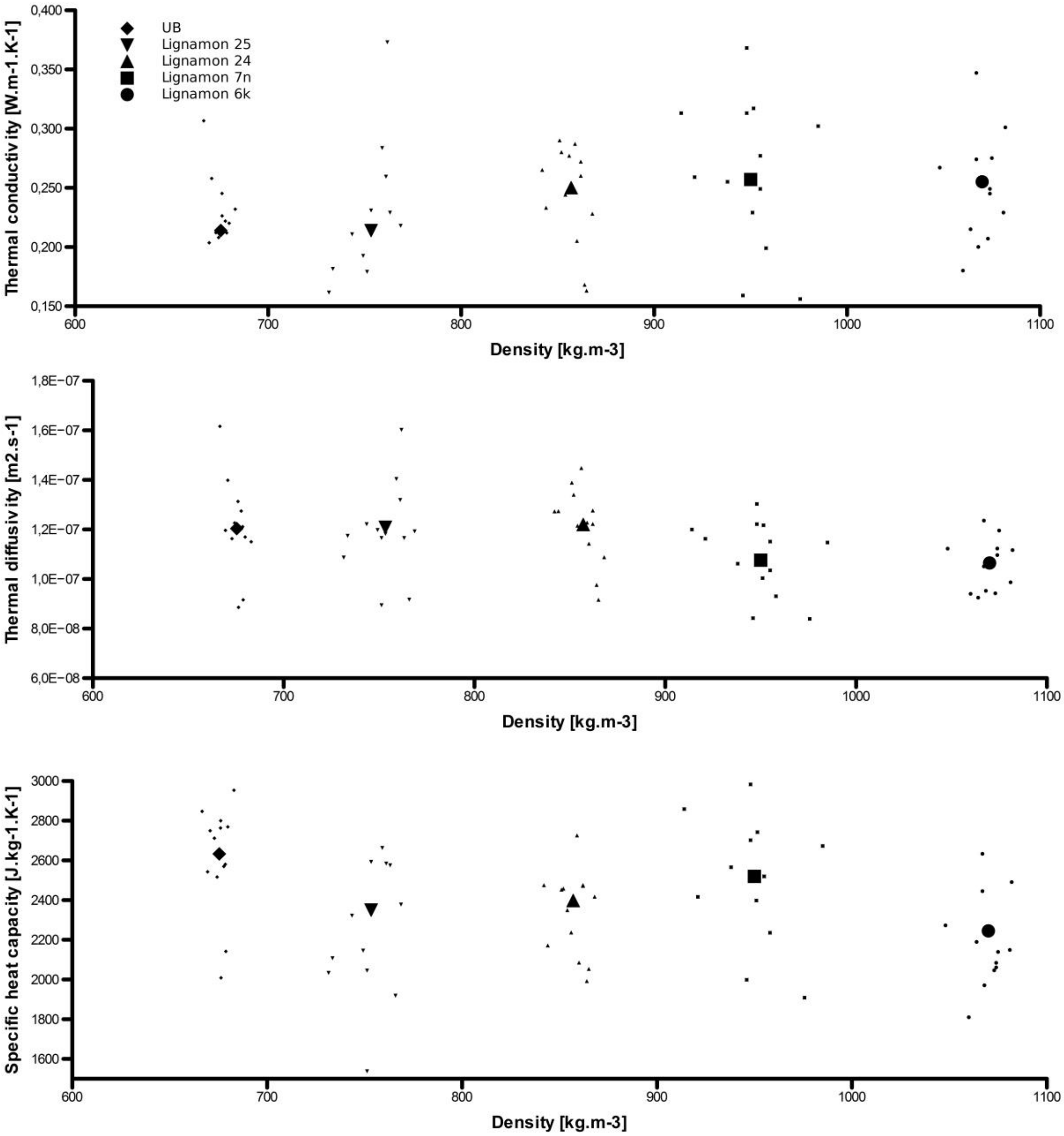

Fig. 5. Dependence of thermal properties on material densities: top: thermal conductivity dependence (λ [W.m^{-1}.K^{-1}]); middle: thermal diffusivity dependence (a [m^2.s^{-1}]); bottom: specific heat capacity dependence (c [J.kg^{-1}.K^{-1}]

The results for untreated beech wood exhibited the lowest value of thermal conductivity (0.21 W.m^{-1}.K^{-1}), but the value of specific heat capacity (2632 J.kg^{-1}K^{-1}) was the highest. Lexa *et al.* (1952) earlier published a thermal conductivity value of 0.19 W.m^{-1}.K^{-1} of *Fagus sylvatica* within the density range of 700 to 1000 kg.m^{-3}. The thermal conductivity (0.18 W.m^{-1}.K^{-1}) of American beech at 12% moisture content with a density of 680 kg.m^{-3} was published also in Simpson and TenWolde (1999). The value of 0.173 W.m^{-1}.K^{-1} in the radial direction and 0.170 W.m^{-1}.K^{-1} in the tangential direction for *Fagus* with a density of 684 kg.m^{-3} was reported by Požgaj *et al.* (1997). The differences between thermal conductivity values available in the literature and our measured data can be caused by the relatively high variation coefficient (21%) (Fig. 5), primarily resulting from the variability of the density between samples, as well as by the measurement method itself (the influence of heat loss through the lateral surfaces and of a leak between central samples where the sensor was placed), sample preparation methods, or technological process deviations.

CONCLUSIONS

1. The test analysis showed statistically significant differences between the results of all the measured properties of modified and native beech wood. For instance, the thermal conductivity of Lignamon 6k (density 1070 kg.m^{-3}) and 7n (density 950 kg.m^{-3}) was 23.8% higher on average than that of UB (density 675 kg.m^{-3}), which should have a practical impact on the use of modified material, *e.g.*, in the case of flooring and heating.
2. The results confirmed the well-known ascending character of thermal conductivity and a descending one for diffusivity as functions of density in a moist environment, as shown earlier by Požgaj *et al.* (1997).
3. The highest value of thermal conductivity was found in Lignamon 6k (0.26 W.m^{-1}.K^{-1} at 1070 kg.m^{-3}) and in Lignamon 7n (0.26 W.m^{-1}.K^{-1} at 950 kg.m^{-3}). The thermal conductivity of the samples increased with increasing density, representing increasing compression extent of the Lignamon.
4. The lowest value of specific heat capacity (2244 J.kg^{-1}.K^{-1}) was found in Lignamon 6k with the highest thermal conductivity value (0.26 W.m^{-1}.K^{-1} at 1070 kg.m^{-3}). The measurements demonstrated the independency of specific heat capacity on density.
5. A higher value of a measured data variation coefficient (ranging from 8.7 to 31%) was caused by the natural variability of the wood material, and it was increased by the modification processes as well as having a contribution from the measuring method itself.

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